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(54) PERMANENT MAGNET MATERIAL, PERMANENT MAGNET AND MANUFACTURE OF PERMANENT MAGNET

(57)Abstract:
PURPOSE: To improve magnetic characteristics by residual amorphous phase of a quench R-Fe-B permanent magnet.
CONSTITUTION: In quench permanent materials containing one or more rare earth metal elements (R) and B with Fe as principal component, said permanent magnet materials comprise a soft magnetic residual amorphous phase less than 10 area percent, a remainder and a crystalline phase containing an R-Fe-B hard magnetic compound. Powder of these permanent magnetic materials is subjected to plastic working and bulk magnets are produced.

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CLAIMS

[Claim(s)]

[Claim 1] A permanent magnet material characterized by consisting of a crystalline substance phase which Fe is used as a principal component (however, it is less than [91at%]), and the remainder generates by heat treatment substantially including a soft magnetism residual amorphous phase below 10 area % in a quenching permanent magnet material containing one sort or two sorts or more of rare earth elements (an abbreviated name is set to "R") and boron, and contains a R-Fe-B system hard magnetism compound.

[Claim 2] A permanent magnet material according to claim 1 characterized by said crystalline substance phase containing a magnetic matter phase still smaller than width of face of a magnetic domain wall.

[Claim 3] A permanent magnet material according to claim 1 or 2 characterized by for said boron, said rare earth elements, and oxygen condensing, and existing and changing out of said crystalline substance phase in said amorphous phase at high concentration.

[Claim 4] A permanent magnet material according to claim 3 which a ratio of a content of boron contained in said crystalline substance phase and boron contained in said amorphous phase is in the range of 1:1.5-1:7, and a ratio of a content of oxygen contained in said crystalline substance phase and oxygen contained in said amorphous phase is in the range of 1:1.5-1:7, and is characterized by mean particle diameter of a crystalline substance phase being 5-100nm.

[Claim 5] A permanent magnet material given [to claims 2-4] in any 1 term with which a crystalline substance phase of said permanent magnet material consists of alpha-Fe and Fe3 B and Nd2 Fe14B, and an amorphous phase consists of B which is R and 0 - 25at% which is Fe and 5 - 20at% which is 70 - 90at%.

[Claim 6] A presentation expressed by at% of permanent magnet material Fea Rb Bc X d (However, R shows one sort or two sorts or more of elements of the rare earth elements.) X Co, Si, Cu, Ti, Zr, Hf, V, Nb, Ta, Mo, It is expressed. one sort chosen from W, aluminum, Cd, Au, In, Mg, nickel, Pd, Pt, Ru, Sn, and Zn, or two sorts or more of elements -- it is -- A permanent magnet material given [to claims 1-5 characterized by being 40<=a<91, 4.5<=b<=35, 0.5<=c<=30, 0<=d<=5, and 9.5 <=b+c] in any 1 term.

[Claim 7] 65<=a<=90, 4.5<=b<=7.9, 2<=c<=10, 0<=d<=5, a permanent magnet material according to claim 6 that is 10 <=b+c.

[Claim 8] A permanent magnet material given [to claims 1-7 whose rare earth elements are Nd] in any 1 term.

[Claim 9] A permanent magnet characterized by combining a permanent magnet material given [to claims 1-8 which have a powder gestalt] in any 1 term by resin.

[Claim 10] A permanent magnet characterized by sticking by pressure a permanent magnet material given [to claims 1-8 which have a powder gestalt] in any 1 term by plastic working.

[Claim 11] A permanent magnet according to claim 10 characterized by carrying out extrusion and having a magnetic anisotropy in the direction of extrusion.

[Claim 12] A manufacture method of a permanent magnet according to claim 10 which carries out extruding of the end of rapidly solidified powder.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] If this invention is described in more detail about a permanent magnet material, it relates to an iron-rare-earth-elements-boron system quenching permanent magnet material, a permanent magnet, and its manufacture method.

[0002]

[Description of the Prior Art] The Nd-Fe-B system bond magnet which was excellent in magnetic properties about the magnet with which small and lightweight-ization an electric product, for autoparts, etc. are called for is expected. According to JP,4-47024,B, it is Nd15Fe 88B7. The liquid of the 5 yuan system presentation which added Co and Y into the Nd-Fe-B system alloy represented is used as a quenching thin band, and the permanent magnet which solidified with Nylon the powder which ground this next is indicated. Moreover, the magnetic energy of the quenching thin band is maximum energy product (BH) max. It is said that they are more than 17MGOe(s) (135 kJ/m3).

[0003] It is also well-known to heat-treat an amorphous alloy thin band and to deposit a microcrystal by above-shown JP,4-47024,B, and it is specifically Nd11Fe72Co eight B7.5V1.5. It has heat-treated for 10 minutes at 650 degrees C. Maximum energy product (BH) max after heat treatment They are 18MGOe(s) (143 kJ/m3).

[0004] JP,3-52528,B indicates the method of performing annealing so that the hard magnetism fine crystalline substance phase of 20-400nm may deposit the liquid quenching magnet alloy by which a presentation is expressed with Nd0.1-0.5 0.9(TM 0.9-0.995B0.005-0.1)-0.5 (however, TMs are transition metals, such as Fe), and the alloy concerned after liquid quenching. The path below a single magnetic-domain field is also suddenly suggested to this fine crystalline substance phase. Furthermore, (BH) max Nd0.15 (Fe 0.95B0.05) 0.85 were indicated to be About 14 MGOe(s) (111 kJ/m3) are reached.

[0005] succeeding also in the attempt which obtains a permanent magnet by the Nd-Fe-B system presentation of the amount of Nd(s) lower than the above-mentioned technology, for example, heat-treating the amorphous ribbon of 77BNd4 Fe19 presentation is proposed -- **** (J.de Phys., such as R.Coehoorn, C 8 and 1988, and pp 669-670) -- it has the defect which is not sufficiently high.

[0006]

[Problem(s) to be Solved by the Invention] Furthermore, by above-shown JP,3-52528,B and JP,4-47024,B, an amorphous phase is made to remain in a ribbon and raising magnetic properties using a residual amorphous phase is not indicated.

[0008] Although the conventional Nd-Fe-B system quenching magnet is highly efficient, in order for the amount of optimal Nd(s) to be more than 10at%, a price becomes high, and there is no competitive strength in a ferrite magnet in respect of a price. Therefore, the ferrite magnet is still used abundantly at the motor of the industrial device more than a medium size, and the actuator. By the way, for Br, 0.2-0.4T, and Hc are [the common magnet property of a ferrite magnet] 0.13 - 0.26 MA/m and (BH) max. 7 - 36 kJ/m3 It is each within the limits and is remarkably inferior to a rare earth magnet efficiently. Under such the actual condition, in respect of the engine performance, it is a Nd-Fe-B system quenching magnet and below equivalent, and the meaning which offers the magnet which does not become remarkably expensive in respect of [ferrite magnet] a price, and expands a magnetic use is large.

[0009] Furthermore, the conventional Nd-Fe-B system quenching magnet powder is used as a bond magnet, and since processability was not excellent, powder was not able to be combined. So, only in the rate of a resin binder, with the conventional bond magnet, the engine performance of a permanent magnet was low.

[0010]

[Means for Solving the Problem] A permanent magnet material concerning this invention is characterized by consisting of a crystalline substance phase which Fe is used as a principal component (however, it is less than [91at%]), and a soft magnetism residual amorphous phase below 10 area % and the remainder generate by heat treatment substantially in one sort or two sorts or more of rare earth elements (an abbreviated name is set to "R"), and a quenching permanent magnet material containing B, and contains a R-Fe-B system hard magnetism compound. Moreover, a permanent magnet concerning this invention is a bond magnet which combined the above-mentioned permanent magnet material of a powder gestalt by resin, or a bulk magnet stuck by pressure by plastic working.

[0011] A permanent magnet material of this invention consists of amorphous phases which remain without crystallizing with a crystalline substance phase which an amorphous alloy crystallizes by heat treatment and generates the organization in a liquid quenching material in which hard magnetism is shown by using Fe, one sort, or two sorts or more of rare earth elements, and B as a principal component. In addition, although the whole organization before heat treatment should just be an amorphous phase preferably, a crystalline substance phase may be contained in the range which does not affect magnetic properties. A R-Fe-B system compound with hard magnetism is contained in a crystalline substance phase. Although there are few places which contribute to improvement in magnetic properties rather than a crystalline substance phase, while an amorphous phase's having soft magnetism on the other hand, and controlling grain growth in the case of heat-of-crystallization processing, enabling formation of a detailed crystalline substance phase and a property as the hard magnetic substance improving, the plastic-working nature of a magnet material is raised. Particle size with this desirable crystalline substance phase is 5-100nm. If particle size is larger than 100nm, a magnetic domain will be formed in an elasticity magnetism phase, and lowering of a residual magnetic flux density Br will be caused. Moreover, since the magnetic properties of a crystalline substance phase will deteriorate if particle size is smaller than 5nm, it is not desirable. A more desirable particle size is 20-50nm.

[0012] On the other hand, if a residual amorphous phase increases more than 10 area %, in order that magnetic association of a crystalline substance interphase may go out, magnetic properties deteriorate. A rate of a desirable residual amorphous phase is two to 10 area %. Furthermore, a desirable range is two to 5 area %. In order to discover work of the above residual amorphous phases, it is desirable to carry out heat treatment temperature to beyond crystallization temperature, and to make heat treatment time amount less than into a short time, i.e., several minutes.

[0013] Furthermore, if said crystalline substance phase contains a soft magnetism matter phase with a size smaller than width of face of a magnetic domain wall, deterioration of a magnet property by elasticity magnetism phase can be controlled. That is, if an elasticity magnetism phase becomes sufficiently smaller than width of face of a magnetic domain wall by existence of a residual amorphous phase, the whole system which magnetization of an elasticity magnetism phase is restrained sufficiently strongly by association with magnetization of a surrounding hard magnetism phase, and becomes from a compound phase by it will act as the one hard magnetic substance, and, as a result, residual magnetic flux density (Br) / coercive force (iHc) will become high.

[0014] In this invention, it is desirable that B, rare earth elements, and oxygen (O) condense and exist rather than a crystalline substance phase in an amorphous phase at high concentration. While making low concentration, such as crystalline substance Aiuchi's B, and Nd, by making suitable heat treatment conditions at the time of crystallizing a part of amorphous phase, B in an amorphous phase and concentration of Nd can be relatively made high. By performing such concentration adjustment, Curie point Tc of an amorphous phase becomes high, magnetic properties in a room temperature improve, and the temperature characteristic of magnetic properties also improves again.

[0015] Moreover, in the above, a content ratio (Oc:Oa) of oxygen (Oc) contained in a crystalline substance phase and oxygen (Oa) contained in an amorphous phase is in the range of 1:1.5-1:7. A content ratio (Bc:Ba) of boron (Bc) contained in a crystalline substance phase and boron (Ba) contained the inside of amorphous is in the range of 1:1.5-1:7, and it is desirable that mean particle diameter of a crystalline substance phase is 5-100nm. Here, Curie point Tc of an amorphous phase will become it low that it is Oa/Oc <1.5, and magnetic properties in a room temperature will deteriorate, and the temperature characteristic of magnetic properties will also deteriorate, and an amorphous phase will make it nonmagnetic that it is Oa/Oc >7. Moreover, amorphous Curie point Tc will become low with it being Ba/Bc <1.5, and an amorphous phase will make it nonmagnetic that it is Ba/Bc >7 too.
[0016] In one embodiment of this invention, a constituent of a crystalline substance phase of a permanent magnet material consists of alpha-Fe and Fe3 B and Nd2 Fe14B, and these crystals constitute mixed crystal. Similarly, an amorphous phase consists of a 70 - 90at% Fe and 5 - 20at% R and 0 - 25at% B.
[0017] A presentation of a permanent magnet material is Fea Rb Bc Xd (however, R shows one sort or two sorts or more of elements of the rare earth elements). X Co, Si, Cu, Ti, Zr, Hf, V, Nb, Ta, Mo, They are one sort or two sorts or more of elements chosen from W, aluminum, Cd, Au, In, Mg, nickel, Pd, Pt, Ru, Sn, and Zn. A magnet property and plastic-working nature which were excellent by filling 9.5at%<=b+c 0 <=d<=5at% 40at%<=a<91at%, 4.5at%<=b<=35at%, and 0.5at%<=c<=30at% can be attained. The inside B of these elements, in order to form an amorphous phase, more than 0.5at% is required, but if it increases more than 30at%, a diameter of crystal grain will become larger than 100nm, and plastic-working nature will fall. Moreover, R and B are required in order that that the total quantity is more than 9.5at% may form an amorphous phase. In addition, an element of X component makes a diameter of crystal grain detailed further, has the role which improves thermal resistance, and dissolves in part in above-mentioned alpha-Fe, Fe3 B, and Nd2 Fe14B, or forms a separate phase.
[0018] A more desirable presentation is 10at%<=b+c 0 <=d<=5at% 65at%<=a<=90at%, 4.5at%<=b<=7.9at%, and 2at%<=c<=10at% from a field of a magnet property. Moreover, a desirable presentation is d<=3at% 78at%<=a<=91at%, 6at%<=b<=12at%, and 3at%<=c<=10at% from a field of plastic-working nature.
[0019] Permanent magnet material powder of this invention can be used as a bond magnet combined with resin, such as Nylon. As a bond magnet, a rate of permanent magnet powder has 95 - 98% of the weight of a common thing. Furthermore, a bulk magnet joins the plastic deformation sides of a powder particle by carrying out plastic deformation of the permanent magnet powder by which rapid solidification was carried out by method of extrusion, a hotpress, etc. However, it is necessary to choose temperature of plastic working so that complete crystallization may not take place. Thus, density of an obtained bulk magnet is usually 99.5% or less of thing to true density.
[0020] When a conjugation method is based on extrusion, extrusion is not made in extrusion temperature of less than 350 degrees C, and in 700-degree-C **, sufficient magnet property is not acquired by big and rough-ization of crystal grain. Extrusion of the reduction of area with which association of powder does not fully accomplish at less than 30%, but exceeds 80% is still more difficult the reduction of area with this alloy. After performing extrusion by the above-mentioned 350-700-degree-C temperature and 30 - 80% of reduction of area, a part of quenching amorphous phase is decomposed into a crystalline substance phase by heat-treating as occasion demands. However, if it exceeds 460 degrees C also by this method, since a quenching amorphous phase will decompose during plastic deformation, heat treatment may be omissible.
[0021] Extrusion temperature and the reduction of area are changed for four kinds of plastic Nd-Fe-B system permanent magnets, extrusion is performed, and a result of having investigated a moldability and a degree of hardness of extrudate is shown in drawing 1 (table 1). In addition, the following criteria estimated a moldability.
O Good pore -- few (99% or more of densities)
O Good ** failure with some pores x with much pore Improper It cannot fabricate, but Fe77Nd 4.5B15 of a moldability is the poorest of these four sorts of the magnet presentations, and other magnet presentations have a moldability in almost equivalent and good level. Moreover, 400-450 degrees C of working temperature are desirable in respect of a moldability.

[0022] Although a powdered bond magnet is isotropy, in this invention, an anisotropy bulk magnet with a high maximum energy product is obtained. for this reason -- being alike -- the easy magnetization direction of a crystal phase carries out orientation to the direction of extrusion by making the reduction of area in extrusion into 70% or more dramatically highly. About an anisotropy, it is the anisotropy energy EA. It measured and it was checked that it is EA !=0. Hereafter, an example explains this invention in detail.
[0023]
[Example] Weighing capacity of Metal Fe, Metal Nd, and the element B was carried out so that it might become the presentation (at%) of a table 2 of No.1-15, and the alloy was produced with the single roll melt quenching method. That is, from the nozzle arranged in the one revolving copper roll upper part, the dissolution metal of the above-mentioned presentation was gushed on said roll with the pressure of argon gas, it quenched, and the thin band was produced. The width of face of the thin band produced as mentioned above was about 2mm, and thickness was about 30 micrometers.
[0024]
[A table 2]
Fe Nd B No.1 90 7 3 No.2 88 7 5 No.3 80 5 15 No.4 85 5 10 No.5 90 5 5 No.6 885 7No.7 89 5 6 No.8* 917 2No.9 90 8 2 No.10 909 1No.11 89 9 2 No.12 8810 2 No.13 89 10 1 No.14 906 4 No.15 89 6 5 Remark: Example of *-comparison [0025] After carrying out heat treatment for the obtained thin band for 3 minutes at 700 degrees C, the result of having investigated the configuration according to the X diffraction is shown in drawing 2 . Bordering on about 10 at(s)%, when larger than this, it became amorphous, and when there was little sum of a presentation of Nd and B, it became the mixed phase of an amorphous substance and a detailed crystal. Therefore, the sum of the amount of Nd and B of the amorphous formation range in Fe-Nd-B the system of 3 yuan is more than 9.5at% at least.
[0026] The result of having investigated the structure in a heat treatment process using DSC and an X diffraction is shown in drawing 3 . In sample No.1 and No.2, three steps of exothermic peaks were observed clearly, and each peak supported crystallization of alpha-Fe and Fe3 B and Nd2 Fe14B. It turns out that sample No.1 in which Fe3 B and an exothermic peak have appeared, and No.2 are excellent in a magnetic property so that it may mention later.
[0027] Next, the alloy thin band of No.1 was heat-treated for 3 minutes at 700 degrees C. The organization of the sample after heat treatment is an alpha-Fe phase, Nd2 Fe14 B phase, and Fe3. He was the mixed phase organization which consists of a detailed crystal which consists of a B phase, and an amorphous phase. Drawing 4 is the TEM photograph which heat-treated sample No.1 of this invention for 3 minutes at 700 degrees C. When this photograph is seen, it turns out that the detailed crystalline substance phase and detailed amorphous phase of about 20-50nm are intermingled.
[0028] Similarly, the result of having measured the presentation of the crystalline substance phase after heat treatment of this invention of sample No.1 and an amorphous phase by three places using EDS is shown in a table 3. It turns out that Nd in amorphous exists mostly compared with Nd in a crystalline substance phase. In addition, the presentation of B was computed from the presentation ratio of the crystalline substance phase and amorphous phase which are shown in a charge presentation and table 5 of B.
[0029]
[A table 3]
The chemical analysis of the configuration phase of sample No.1 (at%) Fe Nd B Analysis (1) Crystalline substance phase 95.3 1.8 2.9 An amorphous phase 83.6 11.2 5.2 Analysis (2) Crystalline substance phase 95.7 1.52.8 Amorphous phase 83.8 9.2 7.0 Analysis (3) Crystalline substance phase 96.9 0.6 2.5 amorphous phase 73.7 13.2 13.1 [0030] Moreover, the result of having measured O and B which are contained in the crystalline substance phase and amorphous phase after sample No.1 of this invention and heat treatment of No.9 using EDS and PEELS, respectively is shown in a table 4. The amount of O in an amorphous phase was 2.6 to 6.5 times the amount of O in a crystalline substance phase, and the amount of B in an amorphous phase was 1.8 to 5.7 times the amount of B in a crystalline substance phase.
[0031]

[A table 4]
· Crystalline substance phase : B in an amorphous phase, the content ratio of O element Sample No.1 Sample No.9 B O B O (1) 1:1.8 1:2.6 1:2.3 1:4.5 (2) 1:2.5 1:2.9 1:4.2 1:5.3 (3) 1:5.31:3.1 1:5.7 1:6.5 [0032] The result of having measured the magnetic properties after heat-treating to a table 5 using VSM is shown. According to the table 5, sample No.1 which is the presentation range of this invention, and No.2 are (BH) max. It excels. it -- receiving -- Fe content -- the presentation of this invention -- sample No.8 out of range -- (BH) max It turns out that it is inferior.
[0033]
[A table 5]
Heat treatment conditions Br Hc (BH) max (T) (MA/m) (kJ/m3) 1 For 700 degrees C and 3 minutes 0.97 0.21 72.0 2 For 700 degrees C and 3 minutes 0.96 0.19 61.4 8 For 700 degrees C and 3 minutes 0.62 0.10 19.7 9 For 700 degrees C and 3 minutes 0.86 0.16 35.7 [0034] It is (BH) max after a heat treatment condition to drawing 2 . The result plotted to the presentation of Nd and B is shown. The total quantity of Nd (rare earth elements) and B is 20 kJ/m3. (BH) max of a more than They are 60 kJ/m3 by carrying out to more than 9.5at%, in order to obtain carrying out to more than 10at% desirable still more preferably. (BH) max It turns out that the thin band which it has is obtained. Therefore, the organization after heat treatment consists of a fine crystal and an amorphous phase, and if Nd, O, and B have condensed at high concentration compared with a crystalline substance phase to this amorphous phase, good magnetic properties will be acquired.
[0035] The quenching thin band of the presentation which consists of Fe89Nd7 B4 by the example 2 single rolling method (at%) was produced. This thin band was amorphous single phase by the X diffraction result. Amorphous powder with a particle size of 150 micrometers or less was obtained by grinding this thin band with a rotor speed mill. The container made from a steel plate (SS41) was filled up with the obtained powder, extrusion was carried out so that it might come after a heat deairing at 300 degrees C and might come 50% of reduction of area at 450 degrees C into a vacuum, and the bulk object of 99% of pack density was acquired. Bulk object tissue was amorphous single phase. In the 1x10-4 or less torrs ambient atmosphere, heat treatment for 5 minutes was performed for the bulk object at 700 degrees C after that. The tissue of the bulk object after heat treatment consisted of amorphous phase, bcc-Fe, and Nd2 Fe14B. Moreover, the bulk object showed the magnet property of Br=13.0kG (1.03 MA/m), iHc=3.2kOe (0.25 MA/m), and (BH) max =14.2 MG-Oe (113 kJ/m3).
[0036] The quenching thin band of the presentation (No.1 of a table 2) which consists of Fe90Nd seven B3 by the example 3 single rolling method (at%) was produced. This thin band was amorphous single phase by the X diffraction result. Amorphous powder with a particle size of 150 micrometers or less was obtained by grinding this thin band with a rotor speed mill. The container made from SS41 was filled up with the obtained powder, extrusion was carried out so that it might come after a heat deairing at 300 degrees C and might come 50% of reduction of area at 490 degrees C into a vacuum, and the bulk object of 99% of pack density was acquired. Bulk object tissue was the mixed phase of an amorphous phase and bcc-Fe. In the 1x10-4 or less torrs ambient atmosphere, heat treatment for 3 minutes was performed for the bulk object at 750 degrees C after that. The tissue of the bulk object after heat treatment consisted of amorphous phase, bcc-Fe, and Nd2 Fe14B. The magnet property of Br=10.8kG (0.86 MA/m), iHc=2.3kOe (0.18 MA/m), and (BH) max =8.8 MG-Oe (70 kJ/m3) was shown. This magnet property showed some lowering as compared with 1 of a table 5.
[0037] The quenching thin band of the presentation which consists of Fe89Nd7 B4 by the example 4 single rolling method (at%) was produced. Amorphous powder with a particle size of 150 micrometers or less was obtained by grinding this thin band with a rotor speed mill. The container of a steel plate (product made from SS41) was filled up with the obtained powder, extrusion was carried out so that it might come after a heat deairing at 300 degrees C and might come 80% of reduction of area at 680 degrees C into a vacuum, the crystal was deposited in processing and coincidence, and the bulk object of 99% of pack density was acquired. The tissue of the bulk object after extrusion consisted of amorphous phase, bcc-Fe, and Nd2 Fe14B. As a result of investigating the magnetic properties of this bulk object, the magnet property of

Br=12.1kG (0.96 MA/m), iHc=2.5kOe (0.19 MA/m), and (BH) max =13.0 MG-Oe (103 kJ/m3) was shown, and the maximum energy product equivalent to a Nd=15at% Nd-Fe-B system quenching thin band was obtained.

[0038]

[Function and Effect of the Invention] In order to make high energy of an amorphous phase / crystalline substance phase-boundaries side, a residual amorphous phase controls grain growth and its magnet property improves (claim 1). Since the method of improving a magnet property using a residual amorphous phase differs from the method of improving the property of the conventional R-Fe-B system compound, it will be able to perform material development in respect of the improvement in magnetic properties of a residual amorphous phase from now on.

[0039] Moreover, by making it detailed, alpha-Fe and Fe3 B which is the elasticity magnetism phase which exists besides Nd2 Fe14B which is a hard magnetism compound in a crystalline substance phase makes these phases a hard magnetism phase, carries out behavior, and raises a magnet property as a result (claims 2 and 5). For this reason, the permanent magnet of the presentation with much Fe in which alpha-Fe is easy to be formed can be manufactured.

[0040] Adoption of the means (claims 3 and 4) which makes Curie point Tc of an amorphous phase high improves the magnetic temperature characteristic.

[0041] If the amount of rare earth elements is lessened (claim 7), it is dramatically advantageous in respect of raw material cost. And the magnet material of this presentation is (BH) max as compared with a ferrite. It is more than an EQC and the application to the device used at the motor for industrial use which Br exceeded the ferrite, and utilized this property since the temperature characteristic was good, an actuator, especially an elevated temperature is expected.

[0042] The bond magnet (claim 9) which solidified the material powder of this invention by resin is a competitive magnet of positioning in the middle of a rare earth magnet and a ferrite magnet conventional in the field of the engine performance and a price. That is, although the engine performance like the former is not required, the magnet of this invention is suitable for a use with the latter dissatisfied engine performance.

[0043] On the other hand, since the magnet engine performance is superior to the bond magnet (claim 9), the bulk magnet (claim 10) which coupled powder directly is a magnet which advanced 1 step in respect of competitive strength. According to this invention, a common anisotropic magnet is obtained in a sintered magnet, without depending on sintering (claim 11). Moreover, since a magnet property equivalent to the conventional quenching thin magnetization stone is acquired even if Nd content decreases a little less than 10% by this, raw material cost reduction has a remarkable thing.

[Translation done.]

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] It is the chart (drawing 1) showing the moldability of four sorts of Nd-Fe-B system magnet powder.
[Drawing 2] The organization of the Nd-Fe-B system magnet relevant to the amount of Nd(s), and the amount of B, and (BH) max It is the shown graph.
[Drawing 3] It is the graph which shows heat treatment temperature and a generation crystalline substance phase.
[Drawing 4] It is the TEM photograph metaloscope (one 2 million times the scale factor of this) which heat-treated sample No.1 of this invention at 700 degrees C for 3 minutes.

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(54)【発明の名称】　永久磁石材料、永久磁石及び永久磁石の製造方法

(57)【要約】

【目的】　急冷Ｒ－Ｆｅ－Ｂ系永久磁石の残留非晶質相により磁石特性を向上させる。

【構成】　Ｆｅを主成分とし、１種または２種以上の希土類元素（Ｒ）とＢを含有する急冷永久磁石材料において、１０面積％以下の軟磁性残留非晶質相と、残部、熱処理により生成しかつＲ－Ｆｅ－Ｂ系硬質磁性化合物を含む結晶質相からなる永久磁石材料。この永久磁石材料粉末を塑性加工してバルク磁石とする。

表１

組　成	押出し温度	減面率	成形性	ビッカース硬度
$Fe_{88}Nd_{7}B_{5}$	３００℃	３０％	△	－
	３５０	３０	○	－
	３８０	５０	○	５５０
	３８０	９０	×	－
	４００	２０	×	－
	４００	２５	△	－
	４００	５０	○	５６０
	４２３	５０	○	５４０
	４５０	５０	○	５３０
$Fe_{89}Nd_{7}B_{4}$	４００	５０	○	５６０
	４００	９０	×	－
	４２５	５０	○	５７０
	４３０	５０	○	－
	４５０	５０	○	－
$Fe_{86}Nd_{8}B_{6}$	４００	５０	○	５５０
	４２５	５０	○	－
	４５０	５０	○	－
$Fe_{77}Nd_{4.5}B_{18.5}$	３５０	３０	×	－
	３５０	５０	×	－
	３８０	５０	×	－
	４２５	５０	△	－
	４５０	５０	△	－

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【特許請求の範囲】

【請求項１】　Ｆｅを主成分（但し９１ａｔ％未満である）とし、１種又は２種以上の希土類元素（略称を「Ｒ」とする）とホウ素を含有する急冷永久磁石材料において、１０面積％以下の軟磁性残留非晶質相を含み、残部が実質的に、熱処理により生成しかつＲ－Ｆｅ－Ｂ系硬質磁性化合物を含む結晶質相からなることを特徴とする永久磁石材料。

【請求項２】　前記結晶質相がさらに磁壁の幅より小さい磁性物質相を含むことを特徴とする請求項１記載の永久磁石材料。

【請求項３】　前記非晶質相中に前記ホウ素、前記希土類元素ならびに酸素が前記結晶質相中よりも高濃度に濃縮して存在して成ることを特徴とする請求項１又は２記載の永久磁石材料。

【請求項４】　前記結晶質相中に含まれるホウ素と前記非晶質相中に含まれるホウ素の含有量の比率が１：１．５～１：７の範囲にあり、前記結晶質相中に含まれる酸素と前記非晶質相中に含まれる酸素の含有量の比率が１：１．５～１：７の範囲にあり、かつ結晶質相の平均粒径が５～１００ｎｍであることを特徴とする請求項３記載の永久磁石材料。

【請求項５】　前記永久磁石材料の結晶質相がα－Ｆｅ、Fe_3B及び$Nd_2Fe_{14}B$からなり、非晶質相が７０～９０ａｔ％のＦｅ，５～２０ａｔ％のR及び0～２５ａｔ％のＢよりなる請求項２から４までの何れか１項記載の永久磁石材料。

【請求項６】　永久磁石材料のａｔ％により表される組成が、$Fe_a R_b B_c X_d$（ただし、Ｒは希土類元素のうちの１種又は２種以上の元素を示し、ＸはＣｏ、Ｓｉ、Ｃｕ、Ｔｉ、Ｚｒ、Ｈｆ、Ｖ、Ｎｂ、Ｔａ、Ｍｏ、Ｗ、Ａｌ、Ｃｄ、Ａｕ、Ｉｎ、Ｍｇ、Ｎｉ、Ｐｄ、Ｐｔ、Ｒｕ、Ｓｎ及びＺｎから選択される１種又は２種以上の元素である）で表され、４０≦ａ＜９１、４．５≦ｂ≦３５、０．５≦ｃ≦３０、０≦ｄ≦５、９．５≦ｂ＋ｃ）であることを特徴とする請求項１から５までの何れか１項記載の永久磁石材料。

【請求項７】　６５≦ａ≦９０、４．５≦ｂ≦７．９、２≦ｃ≦１０、０≦ｄ≦５、１０≦ｂ＋ｃである請求項６記載の永久磁石材料。

【請求項８】　希土類元素がＮｄである請求項１から７までの何れか１項記載の永久磁石材料。

【請求項９】　粉末形態を有する請求項１から８までの何れか１項記載の永久磁石材料を樹脂で結合したことを特徴とする永久磁石。

【請求項１０】　粉末形態を有する請求項１から８までの何れか１項記載の永久磁石材料を塑性加工で圧着したことを特徴とする永久磁石。

【請求項１１】　押出加工されており、かつ押出方向に磁気異方性を有することを特徴とする請求項１０記載の永久磁石。

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【請求項１２】　急冷凝固粉末を押出し加工する請求項１０記載の永久磁石の製造方法。

【発明の詳細な説明】

【０００１】

【産業上の利用分野】本発明は、永久磁石材料に関するものであり、さらに詳しく述べるならば、鉄－希土類元素－ホウ素系急冷永久磁石材料ならびに永久磁石及びその製造方法に関するものである。

【０００２】

【従来の技術】電気製品、自動車部品用などの小型・軽量化が求められる磁石に関しては磁気特性の優れたＮｄ－Ｆｅ－Ｂ系ボンド磁石が期待されている。特公平４－４７０２４号公報によると、$Nd_{15}Fe_{88}B_7$に代表されるＮｄ－Ｆｅ－Ｂ系合金にＣｏとＹを添加した五元系組成の液体を急冷薄帯とし、次にこれを粉砕した粉末をナイロン樹脂で固化した永久磁石が開示されている。又、その急冷薄帯の磁気エネルギは最大エネルギ積（ＢＨ）$_{max}$で１７ＭＧＯｅ（１３５ｋＪ／m^3）以上であると述べられている。

【０００３】前掲特公平４－４７０２４号公報により、非晶質合金薄帯を熱処理して微結晶を析出させることも公知であり、具体的には$Nd_{11}Fe_{72}Co_8B_{7.5}V_{1.5}$を６５０℃で１０分間熱処理している。熱処理後の最大エネルギ積（ＢＨ）$_{max}$は１８ＭＧＯｅ（１４３ｋＪ／m^3）である。

【０００４】特公平３－５２５２８号公報は、$Nd_{0.1-0.5}(TM_{0.9-0.995}B_{0.005-0.1})_{0.9-0.5}$（但しＴＭはＦｅなどの遷移金属である）で組成が表される液体急冷磁石合金、及び当該合金を液体急冷後２０～４００ｎｍの硬質磁性微結晶質相が析出するように焼鈍を行う方法を開示する。この微結晶質相は単一磁区領域以下の径をもつと示唆されている。さらに、$Nd_{0.15}(Fe_{0.95}B_{0.05})_{0.85}$について示された（ＢＨ）$_{max}$は約１４ＭＧＯｅ（１１１ｋＪ／$m^3$）に達している。

【０００５】上記した技術より低いＮｄ量のＮｄ－Ｆｅ－Ｂ系組成で永久磁石を得る試みも為されており、例えば、$Nd_4Fe_{77}B_{19}$組成の非晶質リボンを熱処理することが提案されている（R．Coehoorn等、J. de Phys., C8, 1988, pp669～670）が、キュリー温度が充分高くない欠点を有している。

【０００６】

【発明が解決しようとする課題】さらに、前掲特公平３－５２５２８号及び特公平４－４７０２４号公報では非晶質相をリボン中に残留させ、残留非晶質相を利用して磁気特性を向上させることは記載されていない。

【０００８】従来のＮｄ－Ｆｅ－Ｂ系急冷磁石は高性能であるが、最適Ｎｄ量が１０ａｔ％以上であるためには価格が高くなり、フェライト磁石には価格の面では全く競争力がない。したがってフェライト磁石は、中型以上

の産業機器のモーター、アクチュエーターなどには依然として多用されている。ところで、フェライト磁石の一般的磁石特性はBrが0．2～0．4T，Hcは0．13～0．26MA／m、(BH)$_{max}$は7～36kJ／m^{3}のそれぞれの範囲内であり、性能的には希土類磁石より著しく劣る。このような現状の下では、性能の面ではNd－Fe－B系急冷磁石と同等以下であり、価格の面ではフェライト磁石より著しく高価にならない磁石を提供し、磁石の用途を拡大する意義は大きい。

【0009】さらに、従来のNd－Fe－B系急冷磁石粉末はボンド磁石として使用されており、加工性が優れないために粉末どうしを結合させることができなかった。それ故従来のボンド磁石では樹脂バインダーの割合だけ永久磁石の性能が低くなっていた。

【0010】

【課題を解決するための手段】本発明に係る永久磁石材料は、Feを主成分（但し91at％未満である）とし、1種または2種以上の希土類元素（略称を「R」とする）とBを含有する急冷永久磁石材料において、10面積％以下の軟磁性残留非晶質相と、残部が実質的に熱処理により生成しかつR－Fe－B系硬質磁性化合物を含む結晶質相から成ることを特徴とするものである。また、本発明に係る永久磁石は、粉末形態の上記永久磁石材料を樹脂で結合したボンド磁石、あるいは塑性加工で圧着したバルク磁石である。

【0011】本発明の永久磁石材料は、Fe、1種又は2種以上の希土類元素及びBを主成分として硬質磁性を示す液体急冷材料において、その組織を、熱処理により非晶質合金が結晶化して生成する結晶質相と結晶化しないで残留する非晶質相とから構成したものである。なお、好ましくは熱処理前の組織全体が非晶質相であれば良いが、磁気特性に影響を及ぼさない範囲で結晶質相が含まれていてもよい。結晶質相には硬質磁性をもつR－Fe－B系化合物が含まれている。一方非晶質相は、軟磁性を有しており、結晶質相よりは磁気特性向上に寄与するところは少ないが、結晶化熱処理の際結晶粒の成長を抑制し、微細な結晶質相の形成を可能にし、その硬質磁性体としての特性が向上するとともに、磁石材料の塑性加工性を高める。この結晶質相の好ましい粒径は5～100nmである。粒径が100nmより大きいと軟質磁性相に磁区が形成され、残留磁束密度Brの低下が引き起こされる。また粒径が5nmより小さいと結晶質相の磁気特性が劣化するので好ましくない。より好ましい粒径は20～50nmである。

【0012】一方、残留非晶質相が10面積％より多くなると結晶質相間の磁気的結合が切れてしまうため、磁気特性が劣化する。好ましい残留非晶質相の割合は2～10面積％である。また更に好ましい範囲は2～5面積％である。上記のような残留非晶質相の働きを発現するためには、熱処理温度を結晶化温度以上とし、熱処理時間を短時間すなわち数分以下にすることが望ましい。

【0013】さらに、前記結晶質相が磁壁の幅より寸法が小さい軟磁性物質相を含むと、軟質磁性相による磁石特性の劣化を抑制することができる。すなわち、軟質磁性相が残留非晶質相の存在によって磁壁の幅よりも十分小さくなると、軟質磁性相の磁化が周囲の硬質磁性相の磁化との結合によって十分強く拘束されて複合相からなる系全体が1つの硬質磁性体として振舞い、その結果残留磁束密度（Br）／保磁力（iHc）が高くなる。

【0014】本発明においては、非晶質相中にB、希土類元素ならびに酸素（O）が結晶質相よりも高濃度に濃縮して存在することが好ましい。非晶質相の一部を結晶化する際の熱処理条件を適切にすることによって、結晶質相内のB、Ndなどの濃度を低くする一方、相対的に非晶質相中のB、Ndの濃度を高くすることができる。このような濃度調整を行うことによって、非晶質相のキュリー点Tcが高くなり、室温における磁気特性が向上しまた磁気特性の温度特性も向上する。

【0015】また、上記において、結晶質相中に含まれる酸素（O_c）と非晶質相中に含まれる酸素（O_a）の含有量比率（O_c：O_a）が1：1．5～1：7の範囲にあり、結晶質相中に含まれるホウ素（B_c）と非晶質中に含まれるホウ素（B_a）の含有量比率（B_c：B_a）が1：1．5～1：7の範囲にあり、かつ結晶質相の平均粒径が5～100nmであることが好ましい。ここで、$O_a/O_c<1.5$であると非晶質相のキュリー点Tcが低くなり、室温における磁気特性が劣化し、また磁気特性の温度特性も劣化し、$O_a/O_c>7$であると、非晶質相が非磁性化してしまう。また、$B_a/B_c<1.5$であると非晶質のキュリー点Tcが低くなり、$B_a/B_c>7$であると、やはり非晶質相が非磁性化してしまう。

【0016】本発明の一実施態様においては、永久磁石材料の結晶質相の構成物質がα－Fe、Fe_3B及び$Nd_2Fe_{14}B$からなり、これらの結晶は混晶を構成している。同様に、非晶質相は70～90at％のFe，5～20at％のR及び0～25at％のBよりなる。

【0017】永久磁石材料の組成は、$Fe_a R_b B_c X_d$（ただし、Rは希土類元素のうちの1種又は2種以上の元素を示し、XはCo、Si、Cu、Ti、Zr、Hf、V、Nb、Ta、Mo、W、Al、Cd、Au、In、Mg、Ni、Pd、Pt、Ru、Sn及びZnから選択される1種又は2種以上の元素であり、40at％≦a＜91at％、4．5at％≦b≦35at％、0．5at％≦c≦30at％、0≦d≦5at％、9．5at％≦b＋cを満たすことにより優れた磁石特性と塑性加工性を達成することができる。これらの元素中Bは非晶質相を形成するために、0．5at％以上が必要であるが、30at％よりも多くなると、結晶粒径が100nmより大きくなりかつ塑性加工性が低下す

る。またRとBは合計量が9．5at％以上であることが非晶質相を形成するために必要である。なお、X成分の元素は結晶粒径をさらに微細化し、耐熱性を向上する役割をもち、また上記したα－Fe、Fe_3B及び$Nd_2Fe_{14}B$中に一部固溶するかあるいは別個の相を形成する。

【0018】磁石特性の面からより好ましい組成は、65at％≦a≦90at％、4．5at％≦b≦7．9at％、2at％≦c≦10at％、0≦d≦5at％、10at％≦b＋cである。又、塑性加工性の面から好ましい組成は、78at％≦a≦91at％、6at％≦b≦12at％、3at％≦c≦10at％、d≦3at％である。

【0019】本発明の永久磁石材料粉末はナイロン樹脂などの樹脂により結合したボンド磁石とすることができる。ボンド磁石としては永久磁石粉末の割合は95～98重量％のものが一般的である。さらに、バルク磁石は、急冷凝固された永久磁石粉末を押出、ホットプレスなどの方法により塑性変形させることによって、粉末粒子の塑性変形面どうしを接合したものである。但し、塑性加工の温度は全面結晶化が起こらないように選択する必要がある。このようにして得られたバルク磁石の密度は通常真密度に対して99．5％以下のものである。

【0020】接合法が押出による場合は押出温度350℃未満では押出ができず、700℃超では結晶粒の粗大化により、十分な磁石特性が得られない。さらに減面率が30％未満では粉末同士の結合が十分に成されず、80％を超える減面率の押出加工は本合金では困難である。上記の350～700℃温度かつ30～80％の減面率で押出を行った後は、必要により熱処理を行うことによって、急冷非晶質相の一部を結晶質相に分解する。但し、この方法でも460℃を超えると塑性変形中に急冷非晶質相が分解するから、熱処理を省略できることも＊ある。

【0021】4種類の塑性のNd－Fe－B系永久磁石を押出温度及び減面率を変えて押出加工を行い、成形性及び押出材の硬度を調べた結果を図1（表1）に示す。なお、成形性は以下の基準で評価した。

◎　良好　気孔少い（密度99％以上）
○　可　気孔が多少ある
△　不可　気孔が多い
×　不可　成形できず

これら4種の磁石組成の中では$Fe_{77}Nd_{4.5}B_{15}$が成形性が最も不良であり、その他の磁石組成は成形性がほぼ同等で良好なレベルにある。又、加工温度は400～450℃が成形性の面で好ましい。

【0022】粉末のボンド磁石は等方性であるが、本発明においては最大エネルギ積が高い異方性バルク磁石が得られる。このためには、押出における減面率を非常に高く例えば70％以上とすることにより、結晶相の容易磁化方向が押出方向に配向する。異方性については、異方性エネルギーE_Aを測定し、$E_A \neq 0$であることが確認された。以下、実施例により本発明を詳しく説明する。

【0023】

【実施例】表2のNo．1～15の組成（at％）となるように金属Fe、金属Nd及び元素Bを秤量し、単ロール液体急冷法により合金を作製した。すなわち、1つの回転している銅製ロール上方に配置されたノズルより上記組成の溶解金属をアルゴンガスの圧力により前記ロール上に噴出させ、急冷して薄帯を作製した。以上のようにして作製した薄帯の幅は約2mmであり、厚さは約30μmであった。

【0024】

【表2】

	Fe	Nd	B
No．1	90	7	3
No．2	88	7	5
No．3	80	5	15
No．4	85	5	10
No．5	90	5	5
No．6	88	5	7
No．7	89	5	6
No．8＊	91	7	2
No．9	90	8	2
No．10	90	9	1
No．11	89	9	2
No．12	88	10	2
No．13	89	10	1
No．14	90	6	4
No．15	89	6	5

備考：＊－比較例

【００２５】得られた薄帯を７００℃で３分間熱処理をした後、Ｘ線回折により構成を調べた結果を図２に示す。ＮｄとＢの組成の和が約１０ａｔ％を境にして、これより大きいと非晶質となり、少ないと非晶質と微細な結晶との混相となった。したがって、Ｆｅ－Ｎｄ－Ｂ３元系における非晶質形成範囲はＮｄとＢの量の和が少なくとも９．５ａｔ％以上である。

【００２６】ＤＳＣとＸ線回折を用いて熱処理過程での構造を調べた結果を図３に示す。試料Ｎｏ．１、Ｎｏ．２では３段の発熱ピークが明瞭に観察され、それぞれのピークがα－Ｆｅ、Ｆｅ$_3$ ＢとＮｄ$_2$ Ｆｅ$_{14}$Ｂの結晶化に対応していた。後述するように、Ｆｅ$_3$ Ｂと発熱ピークが現れている試料Ｎｏ．１、Ｎｏ．２は磁気的特性が優れていることがわかる。

【００２７】次に、Ｎｏ．１の合金薄帯を７００℃で３分間熱処理した。熱処理後の試料の組織は、α－Ｆｅ相，Ｎｄ$_2$ Ｆｅ$_{14}$Ｂ相とＦｅ$_3$ Ｂ相からなる微細な結晶と非晶質相からなる混相組織であった。図４は、本発明の試料Ｎｏ．１を７００℃で３分間熱処理したＴＥＭ写真である。この写真を見ると、約２０～５０ｎｍの微細な結晶質相と非晶質相が混在していることがわかる。

【００２８】同じく、本発明の試料Ｎｏ．１の熱処理後における結晶質相と非晶質相の組成をＥＤＳを用いて３箇所で測定した結果を表３に示す。非晶質中のＮｄは、結晶質相中のＮｄに比べて多く存在しているのが分かる。なお、Ｂの組成はＢの投入組成と表５に示す結晶質相と非晶質相の組成比から算出した。

【００２９】

【表３】

試料Ｎｏ．１の構成相の化学分析値（ａｔ％）

		Ｆｅ	Ｎｄ	Ｂ
分析（１）	結晶質相	９５．３	１．８	２．９
	非晶質相	８３．６	１１．２	５．２
分析（２）	結晶質相	９５．７	１．５	２．８
	非晶質相	８３．８	９．２	７．０
分析（３）	結晶質相	９６．９	０．６	２．５
	非晶質相	７３．７	１３．２	１３．１

【００３０】また、表４に本発明の試料Ｎｏ．１およびＮｏ．９の熱処理後の結晶質相および非晶質相に含まれるＯとＢをそれぞれＥＤＳとＰＥＥＬＳを用いて測定した結果を示す。非晶質相中のＯの量は結晶質相中のＯの量の２．６～６．５倍であり、非晶質相中のＢの量は結晶質相中のＢの量の１．８～５．７倍であった。

【００３１】

【表４】

結晶質相：非晶質相中のＢ、Ｏ元素の含有量比率

	試料Ｎｏ．１		試料Ｎｏ．９	
	Ｂ	Ｏ	Ｂ	Ｏ
（１）	１：１．８	１：２．６	１：２．３	１：４．５
（２）	１：２．５	１：２．９	１：４．２	１：５．３
（３）	１：５．３	１：３．１	１：５．７	１：６．５

【００３２】表５に熱処理後の磁気特性をＶＳＭを用いて測定した結果を示す。表５によると、本発明の組成範囲である試料Ｎｏ．１、Ｎｏ．２は、（ＢＨ）$_{max}$ がすぐれている。それに対して、Ｆｅ含有量が本発明の組成範囲外である試料Ｎｏ．８は（ＢＨ）$_{max}$ が劣っているのが分かる。

【００３３】

【表５】

	熱処理条件	Ｂｒ（Ｔ）	Ｈｃ（ＭＡ／ｍ）	（ＢＨ）$_{max}$（ｋＪ／ｍ^3）
１	７００℃、３分間	０．９７	０．２１	７２．０
２	７００℃、３分間	０．９６	０．１９	６１．４
８	７００℃、３分間	０．６２	０．１０	１９．７
９	７００℃、３分間	０．８６	０．１６	３５．７

【００３４】図２に熱処理条件後の（ＢＨ）$_{max}$ をＮｄとＢの組成に対してプロットした結果を示す。Ｎｄ（希土類元素）とＢの合計量は２０ｋＪ／ｍ^3 以上の（ＢＨ）$_{max}$ を得るためには９．５ａｔ％以上とすることが望ましく、さらに好ましくは、１０ａｔ％以上とすることで６０ｋＪ／ｍ^3 の（ＢＨ）$_{max}$ をもつ薄帯が得られることが分かる。したがって、熱処理後の組織が微細結晶と非晶質相から成り、この非晶質相にＮｄ、Ｏ、Ｂが結晶質相に比べて高濃度に濃縮していれば良好な磁気特性が得られる。

【００３５】実施例２

単ロール法によりＦｅ$_{89}$Ｎｄ$_7$ Ｂ$_4$ （ａｔ％）からなる

組成の急冷薄帯を作製した。X線回折結果によりこの薄帯は非晶質単相であった。この薄帯をロータースピードミルで粉砕することにより１５０μm以下の粒径の非晶質粉末を得た。得られた粉末を鋼板（ＳＳ４１）製の容器に充填し、真空中において３００℃で加熱脱気後、４５０℃で減面率５０％になるように押出加工をし、充填密度９９％のバルク体を得た。バルク体の組織は非晶質単相であった。その後バルク体を1×10^{-4}ｔｏｒｒ以下雰囲気において７００℃で５分間の熱処理を行った。熱処理後のバルク体の組織は非晶質相、ｂｃｃ－Ｆｅ、$Nd_2Fe_{14}B$からなっていた。又、バルク体はＢｒ＝１３．０ｋＧ（１．０３MA／m）、ｉＨｃ＝３．２ｋＯｅ（０．２５MA／m）、（ＢＨ）$_{max}$＝１４．２ＭＧ・Ｏｅ（１１３ｋＪ／m^3）の磁石特性を示した。

【００３６】実施例３
単ロール法により$Fe_{90}Nd_7B_3$（ａｔ％）からなる組成（表２のＮｏ．１）の急冷薄帯を作製した。X線回折結果によりこの薄帯は非晶質単相であった。この薄帯をロータースピードミルで粉砕することにより１５０μm以下の粒径の非晶質粉末を得た。得られた粉末をＳＳ４１製の容器に充填し、真空中において３００℃で加熱脱気後、４９０℃で減面率５０％になるように押出加工をし、充填密度９９％のバルク体を得た。バルク体の組織は非晶質相とｂｃｃ－Ｆｅとの混相であった。その後バルク体を1×10^{-4}ｔｏｒｒ以下雰囲気において７５０℃で３分間の熱処理を行った。熱処理後のバルク体の組織は非晶質相、ｂｃｃ－Ｆｅ、$Nd_2Fe_{14}B$からなっていた。Ｂｒ＝１０．８ｋＧ（０．８６MA／m）、ｉＨｃ＝２．３ｋＯｅ（０．１８MA／m）、（ＢＨ）$_{max}$＝８．８ＭＧ・Ｏｅ（７０ｋＪ／m^3）の磁石特性を示した。この磁石特性は表５の１と比較して若干の低下を示した。

【００３７】実施例４
単ロール法により$Fe_{89}Nd_7B_4$（ａｔ％）からなる組成の急冷薄帯を作製した。この薄帯をロータースピードミルで粉砕することにより１５０μm以下の粒径の非晶質粉末を得た。得られた粉末を鋼板（ＳＳ４１製）の容器に充填し、真空中において３００℃で加熱脱気後、６８０℃で減面率８０％になるように押出加工をし、加工と同時に結晶を析出させ、充填密度９９％のバルク体を得た。押出後のバルク体の組織は非晶質相、ｂｃｃ－Ｆｅ、$Nd_2Fe_{14}B$からなっていた。このバルク体の磁気特性を調べた結果、Ｂｒ＝１２．１ｋＧ（０．９６MA／m）、ｉＨｃ＝２．５ｋＯｅ（０．１９MA／m）、（ＢＨ）$_{max}$＝１３．０ＭＧ・Ｏｅ（１０３ｋＪ／m^3）の磁石特性を示し、Ｎｄ＝１５ａｔ％のＮｄ－Ｆｅ－Ｂ系急冷薄帯に相当する最大エネルギ積が得られた。

【００３８】
【発明の作用及び効果】残留非晶質相は、非晶質相／結晶質相界面のエネルギを高くするために、結晶粒の成長を抑制して磁石特性が向上する（請求項１）。残留非晶質相を利用して磁石特性を向上する方法は従来のＲ－Ｆｅ－Ｂ系化合物の特性を向上する方法とは異なっているために、今後残留非晶質相の磁気特性向上の面で材料開発ができる。

【００３９】又、結晶質相中の硬質磁性化合物である$Nd_2Fe_{14}B$の他に存在する軟質磁性相であるα－Ｆｅ、Fe_3Bなども微細化することにより、これらの相を硬質磁性相として挙動させ、結果として磁石特性を向上させる（請求項２、５）。このためにα－Ｆｅが形成され易いＦｅが多い組成の永久磁石を製造することができる。

【００４０】非晶質相のキュリー点Ｔｃを高くする手段（請求項３、４）を採用すると、磁石の温度特性が改良される。

【００４１】希土類元素の量を少なくすると（請求項７）、原料コストの面で非常に有利である。しかもこの組成の磁石材料はフェライトと比較すると（ＢＨ）$_{max}$が同等以上であり、かつＢｒがフェライトを凌駕しかつ温度特性が良好であるので、この特性を活用した産業用モーター、アクチュエーター、特に高温で使用される機器等への応用が期待される。

【００４２】本発明の材料粉末を樹脂で固化したボンド磁石（請求項９）は、性能及び価格の面で従来の希土類磁石とフェライト磁石の中間の位置付けの競争力のある磁石である。すなわち前者ほどの性能は必要ではないが、後者の性能が不満足な用途に本発明の磁石は好適である。

【００４３】一方、粉末を直接結合したバルク磁石（請求項１０）はボンド磁石（請求項９）よりも磁石性能がすぐれているために、競争力の面で一歩進んだ磁石である。焼結磁石では一般的な異方性磁石が、本発明によると焼結に依らずに得られる（請求項１１）。又これによりＮｄ含有量が１０％弱も低減しても従来の急冷薄帯磁石と同等の磁石特性が得られるために、原料コスト低減は著しいものがある。

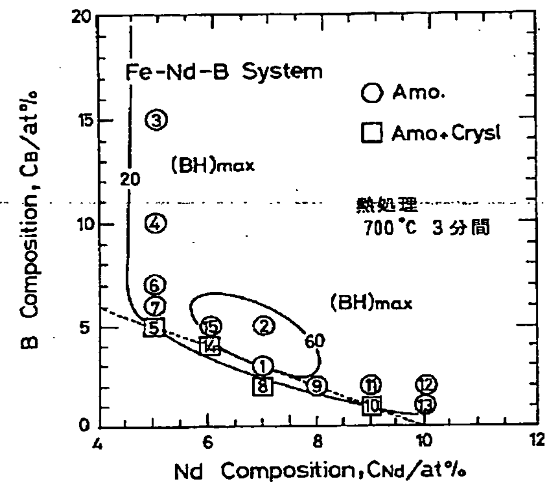
【図面の簡単な説明】
【図１】４種のＮｄ－Ｆｅ－Ｂ系磁石粉末の成形性を示す図表（図１）である。
【図２】Ｎｄ量とＢ量に関連するＮｄ－Ｆｅ－Ｂ系磁石の組織及び（ＢＨ）$_{max}$を示すグラフである。
【図３】熱処理温度と生成結晶質相を示すグラフである。
【図４】本発明の試料Ｎｏ．１を７００℃で３分熱処理したＴＥＭ写真金属顕微鏡（倍率２００万倍）である。

【図1】

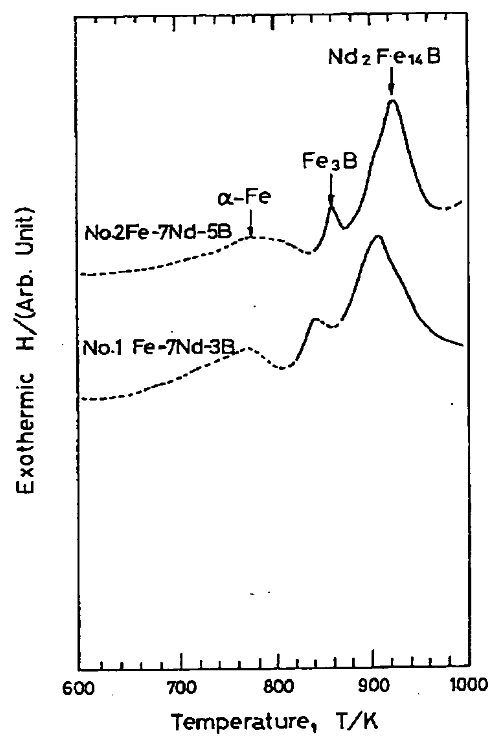
表 1

組 成	押出し温度	減面率	成形性	ビッカース硬度
$Fe_{90}Nd_7B_3$	300℃	30%	△	−
	350	30	○	−
	380	50	○	550
	380	90	×	−
	400	20	×	−
	400	25	△	−
	400	50	○	560
	423	50	○	540
	450	50	○	530
$Fe_{89}Nd_7B_4$	400	50	○	560
	400	90	×	−
	425	50	○	570
	430	50	○	−
	450	50	○	−
$Fe_{89}Nd_8B_3$	400	50	○	550
	425	50	○	−
	450	50	○	−
$Fe_{77}Nd_{4.5}B_{18.5}$	350	30	×	−
	350	50	×	−
	380	50	×	−
	425	50	△	−
	450	50	△	−

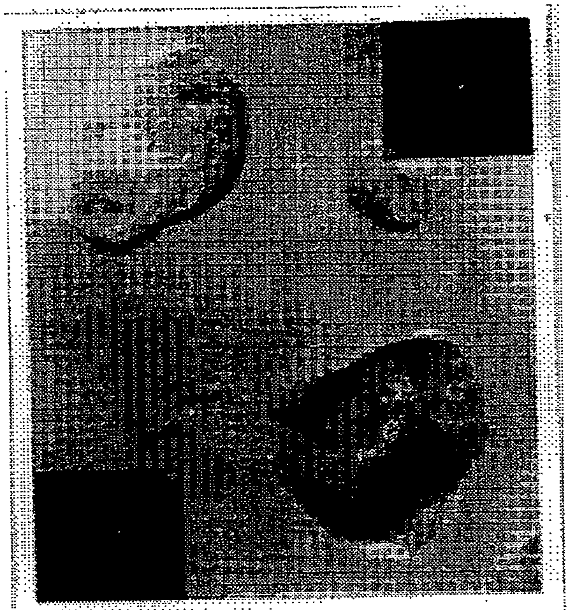
【図2】



【図3】



【図4】



図面代用写真